In-situ characterisation methods of Silicalite-1 formation

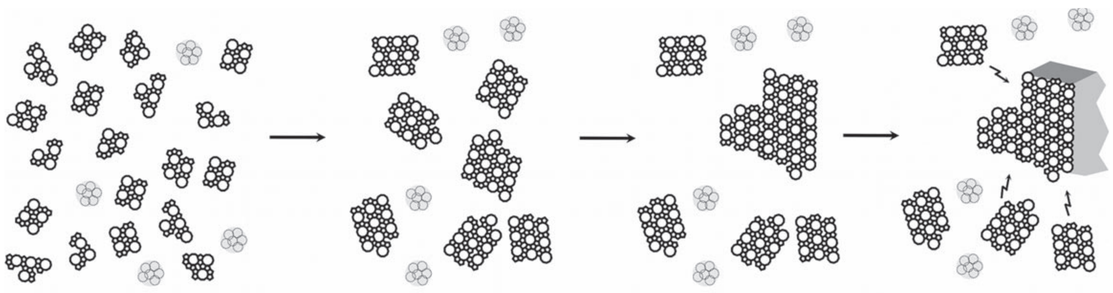
Erwin K. Reichel, Alexander Aerts,
Johan A. Martens, Christine E.A. Kirschhock
Centre for Surface Chemistry and Catalysis (COK)
KU Leuven, Belgium

Abstract

The ongoing research in the field of zeolite (Silicalite-1) formation confirmed the requirement for advanced in-situ characterisation methods of emerging nano-sized crystalline structures.

Applied methods:
- dynamic light scattering (DLS)
- X-ray scattering (SAXS)
- Si NMR
- pH measurement
- rheological parameters
(high-frequency viscosity, loss angle, density)

Synthesis Process



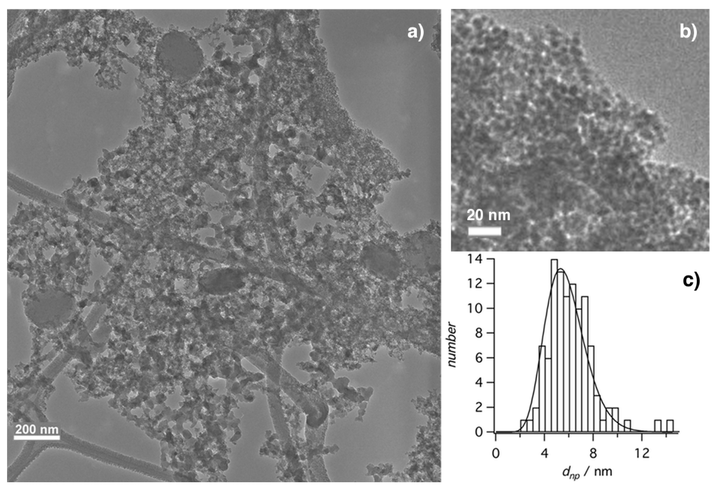
prenucleation *nucleation* *crystal growth*

Clear solution:
- Tetrapropylammonium Hydroxide (TPAOH)
- Tetraethyl orthosilicate (TEOS)
- Water (H_2O)

molar ratios (example): TEOS:TPAOH:H_2O = 25:9:400

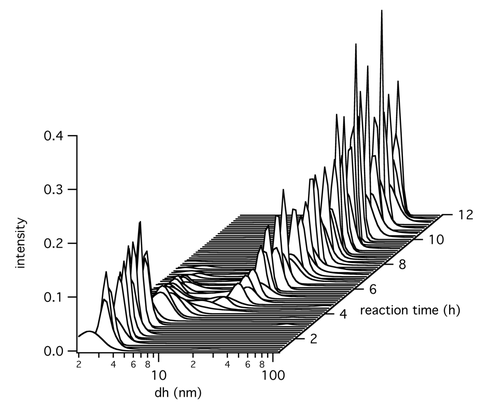
Process conditions:
- temperature: 95°C
- time of synthesis: >20h

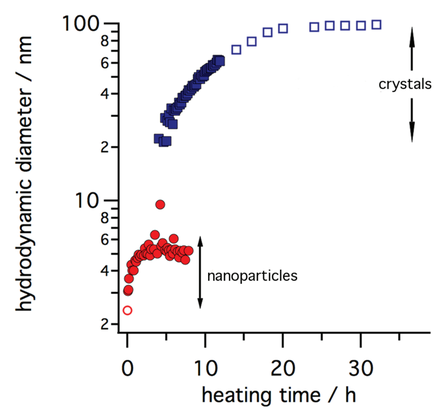
TEM of Nanoparticles and Crystals



erwin.reichel@biw.kuleuven.be
alexander.aerts@biw.kuleuven.be

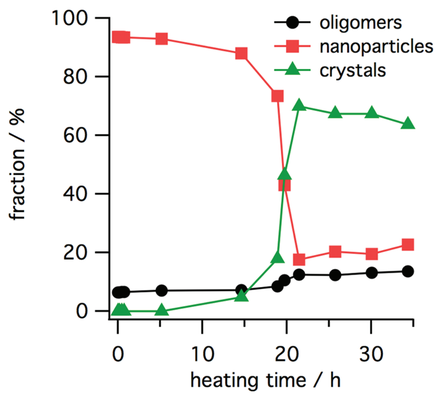
Particle Size: DLS



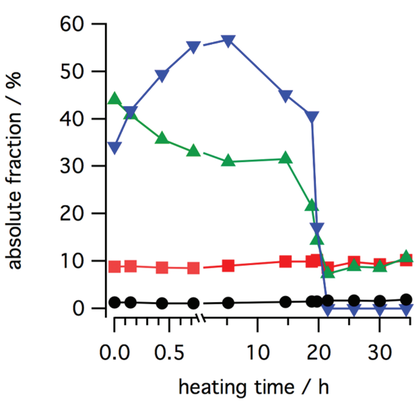


^{29}Si NMR

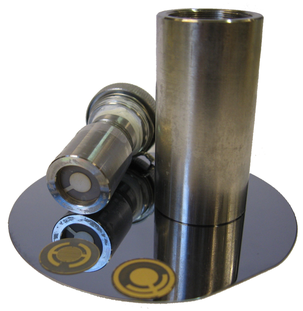
Silica distribution



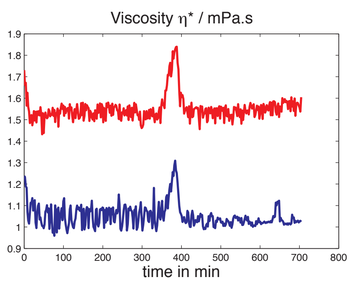
nanoparticle connectivity

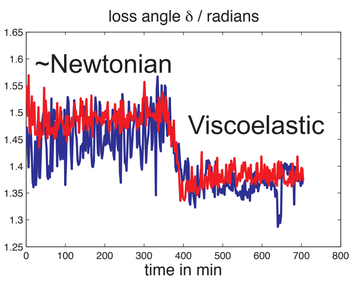


Viscosity sensor



- thickness shear mode (TSM) quartz
- fundamental (4.9 MHz)
- second harmonic (14.7 MHz)





Conclusions

- In-situ diagnostics (DLS, viscosity) provide an efficient way to monitor zeolite synthesis and supplement data from ex-situ analysis (SAXS and Si NMR).
- Viscosity measurement enables accurate calculation of particle size from DLS experiments.

Acknowledgement

FWO, PRODEX, ESA, METHUSALEM, CECAT

[1] Aerts et al, Investigation of the Mechanism of Colloidal Silicalite-1 Crystallization by Using DLS, SAXS, and 29Si NMR Spectroscopy, Chemistry - A European Journal, 16/2010, doi: 10.1002/chem.200901688
[2] Follens et al, Viscosity sensing in heated alkaline zeolite synthesis media, PCCP 11/2009, doi: 10.1039/b816040f